

with respect to the amended claims. Claim 9 is canceled. Claims 10-12 are amended to include the mixed oxide of claim 1.

Rejection of claims 9-14, 19 and 41-42 under 35 U.S.C. §112, second paragraph is respectively traversed with respect to the amended claims. Claims 10-14 are amended. The term densifying temperature is removed from claims 13 and 14. The term "heat treatment temperature" is defined in the application on pages 42, line 21 to page 44, line 4. Basically, it is a method of heat treatment, i.e., sintering the deposited oxide material to provide a material with greater density after heat treatment.

Applicants respectively traverse the rejection to claim 19, and request that this rejection be withdrawn. Applicants argue that one of skilled in the art would understand that the maximum temperature reached during the sintering process of the mixed conducting oxide of the porous body is in a range from 1200°C to 1400°C. Therefore, one would simply heat to a maximum temperature that lies within the claimed range. In other words, the maximum sintering temperature can be 1200°C, 1210°C...1300°C, 1310°C...1400°C. Presenting the claim in this manner is no different than presentation of a Markash group separated by 1°C or 10°C, as indicated above, or claiming deposition of a layer to a specified thickness range.

Rejection of claims 1-9, 11-17, 23-25 and 40 under 35 U.S.C. §103 (a) as being anticipated by Keskar et al. (US 5,964,922) is respectfully traversed. The Examiner rejects claims 1 and 2 of the present invention because Keskar discloses oxides which includes Ta in Table I. Among Table I, mixed conducting solid electrolytes of 7 and 14 include Ta.

The mixed conducting, solid electrolyte 7, as described by the disclosed composition formula, contains a  $\text{Bi}_2\text{O}_3$  base structure. As a result, one of ordinary skill in the art recognizes that solid electrolyte 7 does not include a perovskite type crystal structure, as claimed. Accordingly, applicants request that the rejection based on solid electrolyte 7 be withdrawn.

Compound 14, as described by the disclosed composition formula, is a dual phase mixed conductor (electronic/ionic). Compound 14 is also not recognized by one of ordinary skill in the art as a mixed conducting oxide of perovskite type. Moreover, incorporation of Ta is described as providing one example of a high-temperature metallic phase of compound 14. As a result, Ta is not a component of a substitutional solid solution oxide phase, as claimed. Accordingly, applicants request that the rejection based on compound 14 be withdrawn.

For the reasons above, Table I of Keskar, which the Office Action cites as a prior art, describes materials entirely different from a perovskite type mixed conducting oxide including



Nb or Ta of the present invention. Moreover, Keskar does not teach that the incorporation of Nb or Ta into a perovskite type oxide can stabilize such crystal structures, which is one basis of the claimed invention. Accordingly, Applicants request that the rejection is improper based on the teachings of Keskar, and that the rejection be withdrawn.

The Examiner rejects claim 16 of the present invention because the prior art disclosed on Table I, composition 13 and column 10, lines 51-55 of Keskar. The materials for a mixed conducting solid electrolyte are disclosed in a wide range as  $\text{Sr}_x\text{Fe}_y\text{Co}_z\text{O}_w$  and  $(\text{La}_{1-x}\text{Sr}_x)(\text{Co}_{1-y}\text{Fe}_y)\text{O}_w$ , and indeed include composition range of  $\text{AFe}_x\text{O}_{(3-\delta)}$  of formula (2) of claim 16. However, in rejecting claim 16 on this basis alone, the Office Action fails to consider the entirety of the claimed composite structure.

The composition of claim 16 is directed to a composite material for oxygen separation, which includes a mixed conducting oxide of formula (2) as a porous substrate, and a composition by formula (3) as a dense film. With this combination, materials for composition restricted by formula (2) have excellent stability without decomposition even though they are calcined at high-temperature. The mixed conducting oxide includes a porous substrate, of which the porosity is not lowered during the calcination process of forming the dense film. In contrast, materials described by Keskar, by formula (2), crack easily in a calcination process as the dense body is produced. As a result, the described materials of Keskar are not suitable as a dense film material.

The Office Action cites to the oxide formula on Col. 9, line 39, and to the general disclosure that this oxide can also contain one of the listed secondary metals including Ta. However, there is no teaching as to how much of these secondary metals are added to the oxide, just that they can be added. Also, there is no teaching or suggestion in Keskar that Ta is particularly desirable out of the seven secondary metals listed in the reference.

As discussed in the application, Applicants were confronted with the problem of identifying a particular class of perovskite mixed oxide that was stable in spite of a relatively high density of oxygen holes. Page 26, line 17 to page 27, line 11. A relatively high density of oxygen holes typically provides greater oxygen atom transfer rates through the oxide. To solve the instability problem associated with high oxygen hole densities in known oxides, and the ultimate collapse of the porous oxide, Applicants determined that the incorporation of specific amounts of Nb and Ta into the oxide structure can have a significant stabilization effect. The effect observed with Ta or Nb is greater than iron, which is metal typically used for cubic



structure stabilization. Applicants also determined that the amount of Nb or Ta that is incorporated into the oxide structure is important. The optimal range is defined, and claimed, as  $0 < y \leq 0.5$ . In fact, Applicants discovered that “[i]f the amount of Nb and Ta is increased beyond this range, ...the oxygen permeability of the composite material is lowered.” Page 28, lines 13-18.

Because there is no teaching in Keskar that the selection of Ta out of the seven listed secondary metals provides a relatively stable oxide structure with high oxygen hole densities, and that an amount of Ta, as claimed, is required to meet this condition is not taught or suggested by Keskar, the rejection under §103(a) is improper. In summary, because there is no mention of the problem of oxide instability, and that any one of many possible oxide structures recited listed in Table 1 can be used as a porous oxide membrane, there is no teaching or suggestion in Keskar to select Ta out of the list of seven metals, and then moreover, to add Ta to the oxide structure (1) in the amounts claimed. For these reasons the rejection is improper, and Applicants respectfully request that this rejection be withdrawn.

Rejection of claims 35 and 38 under 35 U.S.C. §102(b) as being anticipated by Gottzmann (US 5,820,655) and claims 36-37 under 35 U.S.C. §103(a) as being unpatentable over Gottzmann in view of Keskar is respectfully traversed with respect to the amended claims. Claims 35 was amended to include the limitation to percent porosity. The amendment is supported by the disclosure recited at the top of page 35. Because there is no teaching in Gottzmann of an oxide with a porosity as claimed, Applicants respectfully request that this rejection be withdrawn.

Rejection of claims 18, 20-22, 28-34 and 39 under 35 U.S.C. §103 (a) as being unpatentable over Keskar in view of Gottzmann is respectfully traversed. Claims 18 and 21 were canceled.

With respect to the rejection of claims 20 and 22, which depend on claims 13 and 14, respectively, Applicants respectfully request that this rejection be withdrawn. As stated above, Keskar does not teach or suggest the selective use of Nb or Ta within the claimed range. Also, there is no teaching in Keskar or Gottzmann that the dense continuous layer, or ion transport membrane as referred to in Keskar, be of a specific oxide structure as claimed.

With respect to claims 28-34, Applicants respectfully request that this rejection be withdrawn. Applicants assert that claims 28 and 30-34, which depend directly or indirectly on claim 16, are patentable. Claim 16 was amended, and is in condition of allowance.



Applicants respectfully request that the rejection of claims 36-37 and 41-42 under 35 USC §103(a) as being unpatentable over Gottzmann as applied to claim 35 in view of Keskar be withdrawn. Claim 35 is amended, and for the reasons stated above, claims 36 and 37 are in condition for allowance.

With respect to claims 41 and 42, there is no teaching in either of the cited references that a composite material can be made by forming a gastight continuous layer over a porous body portion by sintering the oxide material used to from the porous body at a maximum sintering temperature greater than the maximum sintering temperature of an oxide material used to make the gastight layer.

In view of the above, each of the presently pending claims in this application is believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to withdraw the outstanding rejection of the claims and to pass this application to issue.

Attached hereto is a marked-up version of the changes made to the abstract and claims by the current amendment. The attached page is captioned "Version with markings to show changes made." Applicant believes no fee is due with this response. However, if a fee is due, please charge our Deposit Account No. 22-0185, under Order No. 21776-055-US from which the undersigned is authorized to draw.

Dated:

Respectfully submitted,

By

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**Version With Markings to Show Changes Made****In the Abstract:**

A ceramic composite [is] with a mixed conducting oxide that has perovskite type crystal structure of  $\{Ln_{1-a}A_a\} \{B_xB'_yB''_z\} O_{(3-\delta)}$  where a, x, y, and z are within the range of  $0.8 \leq a \leq 1$ ,  $0 < x, 0 < y \leq 0.5$ ,  $0 \leq z \leq 0.2$ ,  $0.98 \leq x + y + z \leq 1.02$ , and  $\delta$  denotes a value that is determined so as to meet a charge neutralization condition. [Ln denotes a combination of one or more kinds of elements selected from Y or lanthanide elements;] A denotes a combination of one or more kinds of elements selected out of Ba, Sr, and Ca. B denotes a combination of one or more kinds of elements selected out of Co, Fe, Cr, and Ga, the combination always containing Fe or Co[,]. [wherein a summation of molar numbers of Cr and Ga is 0% to 20% of a total molar number 'x' of element B;] B' denotes a combination of one or more kinds of elements selected out of Nb, Ta, Ti, and Zr, the combination always containing Nb or Ta[,]. [and where a summation of molar numbers is 0% to 20% of a total molar number 'y' of elements B'; and B'' denotes a combination of one or more kinds of elements selected out of Cu, Ni, Zn, Li, and Mg. This ceramic composite has its excellent properties as a dense film as well as a porous support body. In addition, another type of porous support body that the] The present invention [provides is composed of] is also directed to a mixed conducting oxide and a ceramic composite. [ wherein, when the compositional] The mixed conducting oxide is of formula [is expressed as]  $AFe_xO_{(3-\delta)}$ , [ the composite formula is constituted.] A is selected out of Ba, Sr, and Ca, and is within the range of  $0.98 \leq x \leq 1.02$ , and  $\delta$  denotes a value determined so as to meet the charge neutralization conditions.



**In the Claims:**

Please cancel claims 9, 15, 18, and 21.

Please amend claims 10-14, 16, 28, 33-35 and 39-42 as follows:

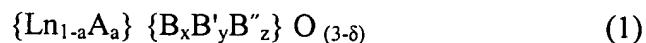
10. (Amended) A method of producing a composite material, wherein a porous body portion comprising a mixed conducting oxide of claim 1 is sintered at a temperature higher than the sintering temperature for a dense continuous layer of a mixed conducting oxide to be formed on said porous body portion, and then a film portion including said dense continuous layer is formed on said porous body portion.

11. (Amended) A composite material comprising a porous body portion comprising a mixed conducting oxide of claim 1, and a film portion including a dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein the densifying temperature for the oxide material of said porous body portion is higher than that for the material of said film portion.

12. (Amended) A material according to claim [9 or ]11, wherein the porosity of said porous body portion is within the range of 20% to 80%, and the thickness of said dense continuous layer is within the range of 10  $\mu\text{m}$  to 1mm.

13. (Amended) A composite material comprising a porous body portion comprising a mixed conducting oxide, and a film portion including a dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein the [densifying temperature for the oxide material of said porous body portion is higher than that for the material of said film portion or the] maximum heat treatment temperature for said porous body portion is higher than that for said dense continuous layer, and

said porous body portion comprises a ceramic composition as a mixed conducting oxide in perovskite structure, said composition being expressed by the following general formula (1):



where Ln represents one or a combination of elements selected from the group of Y and



lanthanoids;

A represents one or a combination of elements selected from the group of Ba, Sr, and Ca;

B represents one or a combination of elements selected from the group of Co, Fe, Cr, and Ga, B always containing Fe or Co, the sum of the molar numbers of Cr and Ga being within the range of 0% to 20% of the total molar number x of B;

B' represents one or a combination of elements selected from the group of Nb, Ta, Ti, and Zr, B' always containing Nb or Ta, the sum of the molar numbers of Ti and Zr being within the range of 0% to 20% of the total molar number y and B';

B" represents one or a combination of elements selected from the group of Cu, Ni, Zn, Li, and Mg;

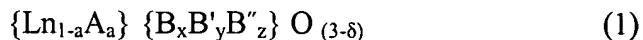
$0.8 \leq a \leq 1$ ;  $0 < x$ ;  $0 < y \leq 0.5$ ;  $0 \leq z \leq 0.2$

$0.98 \leq x + y + z \leq 1.02$ ; and

$\delta$  represents a value which is so determined as to meet charge neutralization conditions.

14. (Amended) A composite material comprising a porous body portion comprising a mixed conducting oxide, and a film portion including a dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein the [densifying temperature for the oxide material of said porous body portion is higher than that for the material of said film portion or the] maximum heat treatment temperature for said oxide material of said porous body portion is higher than that for said dense continuous layer, and

said porous body portion comprises a ceramic composition as a mixed conducting oxide in perovskite structure, said composition being expressed by the following general formula (1):



where Ln represents one or a combination of elements selected from the group of Y and lanthanoids;

A represents one or a combination of elements selected from the group of Ba, Sr, and Ca;

B represents one or a combination of elements selected from the group of Co, Fe, Cr, and Ga, B always containing Fe, the molar number of Co being within the range of 0% to 10% of the total molar number of Fe, the sum of the molar numbers of Cr and Ga being within the range of 0% to 20% of the total molar number x of B;



B' represents one or a combination of elements selected from the group of Nb, Ta, Ti, and Zr, B' always containing Nb or Ta, the sum of the molar numbers of Ti and Zr being within the range of 0% to 20% of the total molar number y and B';

B" represents one or a combination of elements selected from the group of Zn, Li, and Mg;

$0.8 \leq a \leq 1$ ;  $0 < x$ ;  $0 < y \leq 0.5$ ;  $0 \leq z \leq 0.2$

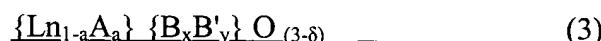
$0.98 \leq x + y + z \leq 1.02$ ; and

$\delta$  represents a value which is so determined as to meet charge neutralization conditions.

16. (Amended) A composite material [comprising] with a porous body portion comprising a mixed conducting oxide, and a film portion including a gastight dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein said [porous body portion comprises a porous body comprising a] mixed conducting oxide of said porous body portion is expressed by [the following] general formula (2):



where  $0.98 \leq x \leq 1.02$ ; A represents one or a combination of elements selected from the group of Ba, Sr, and Ca; and  $\delta$  represents a value which is so determined as to meet charge neutralization conditions and said mixed conducting oxide of said dense continuous layer is general formula (3):



where Ln represents one or a combination of elements selected from the group of Y and lanthanoids;

A represents one or a combination of elements selected from the group of Ba, Sr, and Ca;

B represents one or a combination of elements selected from the group of Fe and Co;

B' represents one or a combination of elements selected from the group of Cu, Ni, Zn, Li, and Mg;

$0.8 \leq a \leq 1$ ;  $0 < x$ ;  $0 \leq y \leq 0.2$ ;

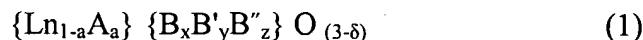
$0.98 \leq x + y \leq 1.02$ ; and

$\delta$  represents a value which is so determined as to meet charge neutralization conditions.

28. (Amended) A composite material [according to claim 16, wherein said] with a dense continuous layer [is made of a ceramic composition as] comprising a mixed conducting



oxide in perovskite structure, said composition being expressed by the following general formula (1)



where Ln represents one or a combination of elements selected from the group of Y and lanthanoids;

A represents one or a combination of elements selected from the group of Ba, Sr, and Ca;

B represents one or a combination of elements selected from the group of Co, Fe, Cr, and Ga, B always containing Fe or Co, the sum of the molar numbers of Cr and Ga being within the range of 0% to 20% of the total molar number x of B;

B' represents one or a combination of elements selected from the group of Nb, Ta, Ti, and Zr, B' always containing Nb or Ta, the sum of the molar numbers of Ti and Zr being within the range of 0% to 20% of the total molar number y and B';

B'' represents one or a combination of elements selected from the group of Cu, Ni, Zn, Li, and Mg;

$$0.8 \leq a \leq 1; 0 < x; 0 < y \leq 0.5; 0 \leq z \leq 0.2$$

$$0.98 \leq x + y + z \leq 1.02; \text{ and}$$

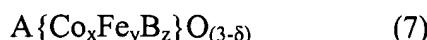
$\delta$  represents a value which is so determined as to meet charge neutralization conditions; and

a porous body portion comprising a mixed conducting oxide, said porous body portion is expressed by general formula (2):



where  $0.98 \leq x \leq 1.02$ ; A represents one or a combination of elements selected from the group of Ba, Sr, and Ca; and  $\delta$  represents a value which is so determined as to meet charge neutralization conditions.

33. (Amended) A material according to claim 13 [or 16], wherein said dense continuous layer is made of a ceramic of a mixed conducting oxide having its composition expressed by the following general formula (7):

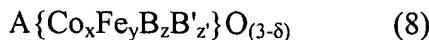


where A represents one or a combination of elements selected from the group of Ba, Sr, and Ca; B represents one or a combination of elements selected from the group of Nb and Ta;



$0 \leq x; 0 \leq y; 0 < z \leq 0.2; 0.98 \leq x + y + z \leq 1.02$ ; and  $\delta$  represents a value which is so determined as to meet charge neutralization conditions.

34. (Amended) A material according to claim 13 [or 16], wherein said dense continuous layer is made of a ceramic of a mixed conducting oxide having its composition expressed by the following general formula (8):



where A represents one or a combination of elements selected from the group of Ba, Sr, and Ca; B represents one or a combination of elements selected from the group of Nb and Ta; B' represents one or a combination of elements selected from the group of Cu, Ni, and Zn;  $0 \leq x; 0 \leq y; 0 < z \leq 0.2; 0 \leq z' \leq 0.2; 0.98 \leq x + y + z + z' \leq 1.02$ ; and  $\delta$  represents a value which is so determined as to meet charge neutralization conditions.

35. (Amended) A composite material wherein an oxygen exchange layer is formed on a surface of one or either side of an oxide having oxide ion diffusivity and porosity from 20% to 80%, said layer being made of an oxide having its composition different from said oxide having oxide ion diffusivity.

39. (Amended) A material according to any of claims 3, 4[, 9, 11, 13, 14, and 20 to 29] or 16, wherein an oxygen exchange layer is formed on a surface of one or either side of said dense continuous layer, said oxygen exchange layer being made of an oxide [having its] of different composition [different from] than the oxide forming said dense continuous layer.

40. (Amended) A material according to any of claims 3, 4[, 13, 14, and 20 to 29,] or 16 wherein the porosity of said porous body portion is within the range of 20% to 80%, and the thickness of said dense continuous layer is within the range of 10  $\mu$ m to 1mm.

41. (Amended) [An oxygen separator including] A method of making a composite material for the separation of oxygen from a mixed gas, comprising: providing a porous body portion comprising a mixed conducting oxide[,]; and providing a film portion including a gastight dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein the maximum heat treatment temperature for said mixed conducting oxide of porous body portion [is] includes sintering at a higher maximum temperature than that of said



dense continuous layer.

42. (Amended) [A chemical reactor including] A method of making a composite material for use as a chemical reactor, comprising: providing a porous body portion comprising a mixed conducting oxide[,]; and providing a film portion including a gastight dense continuous layer of a mixed conducting oxide formed on said porous body portion, wherein the maximum heat treatment temperature for said mixed conducting oxide of porous body portion [is] includes sintering at a higher maximum temperature than that [for] of said dense continuous layer.

